

On-Line Measurement of Heat of Combustion of Gaseous Hydrocarbon Fuel Mixtures

Danny R. Sprinkle
Langley Research Center • Hampton, Virginia

Sushil K. Chaturvedi and Ali Kheireddine
Old Dominion University • Norfolk, Virginia

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Abstract

A method for the on-line measurement of the heat of combustion of gaseous hydrocarbon fuel mixtures has been developed and tested. The method involves combustion of a test gas with a measured quantity of air to achieve a preset concentration of oxygen in the combustion products. This method involves using a controller which maintains the fuel (gas) volumetric flow rate at a level consistent with the desired oxygen concentration in the combustion products. The heat of combustion is determined from a known correlation with the fuel flow rate. An on-line computer accesses the fuel flow data and displays the heat of combustion measurement at desired time intervals. This technique appears to be especially applicable for measuring heats of combustion of hydrocarbon mixtures of unknown composition such as natural gas.

Introduction

Several well-known techniques are available currently for the measurement of heat of combustion of hydrocarbon fuels. Constant-volume bomb calorimeters are commonly used for determining heat of combustion of liquid fuels (refs. 1 and 2). Similarly, constant-pressure flame calorimeters are employed for measuring the heat of combustion of gaseous fuels (refs. 3 and 4). Singh et al. (ref. 5) have developed an on-line technique which requires injection of pure oxygen prior to combustion for measuring the heat of combustion of gaseous fuels. The injection is necessary to maintain the required 21-percent oxygen concentration in the combustion products. Singh et al. have also developed a simplified method that eliminates the need for the addition of pure oxygen and uses a computational algorithm that is valid only for a prescribed air flow rate and combustion-product oxygen concentration (ref. 6). The present technique offers an improvement over this latter method by using a computational algorithm that is valid over a wide range of flow rates and oxygen concentrations. Development of the present measurement system satisfies the need for accurate, on-line measurement of heat of combustion of gaseous fuel mixtures with methane as the dominant component. This fuel has been used in the combustor of the Langley 8 Foot High-Temperature Tunnel (8-Ft HTT) to produce temperature levels approaching 2000 K.

In the present study, an on-line gas calorimeter was developed and tested for high-precision measurement of heat of combustion of gaseous hydrocarbon mixtures. The method described herein involves the measurement of the concentration of oxygen in the combustion products and its correlation with the heat of combustion of the fuel.

Symbols

$$A = 4 \sum_{i=1}^m f_i x_i + (1 + P) \sum_{i=1}^m f_i y_i - 4P$$

C_f	fuel flow rate correction factor
$C_x H_y$	hydrocarbon
E	optical encoder data
f	total hydrocarbon mole fraction
f_i	mole fraction of i th hydrocarbon species
I	inert fraction of fuel
l	air volumetric flow rate
m	number of hydrocarbon species
n	fuel volumetric flow rate
P	combustion-product oxygen mole fraction
P_o	mole fraction of oxygen in air
R^2	coefficient of determination
α, β	regression coefficients
ΔH	heat of combustion

Experimental Setup

The prototype gas calorimeter is shown schematically in figure 1 and photographically in figure 2. The fuel gas is mixed with air and fed into a specially designed burner. The fuel flow rate is measured by a volumetric flowmeter with an optical encoder mounted on its shaft. The flow rate is determined by measuring the counts/min of the etched lines on the encoder disk. Each revolution of the shaft, which is nominally equivalent to 1000 cc, produces 1200 counts.

A burner that is fabricated with an internal honeycomb structure was used to ensure a stable flame over a range of air and fuel flow rates. The burner was enclosed in a vented glass container which prevented atmospheric contamination of the combustion products.

The oxygen concentration in the combustion products was measured by pumping the combustion products through a yttrium-stabilized zirconium oxide oxygen sensor that produces a voltage related to the concentration of oxygen in the environment to which it is exposed (ref. 7). The oxygen sensor output was directed to a

controller that maintained the desired concentration of oxygen in the combustion products by varying the fuel flow rate. This technique requires a sufficient quantity of oxygen (>0.1 percent) in the combustion products to be measurable by the oxygen sensor. A set point concentration of 3-percent oxygen was used throughout this study.

Because of the precision uncertainties of the volumetric flowmeter, the oxygen sensor, and the air flowmeter, and because of the inherent unsteadiness of the combustion process, the oxygen concentration in the product gases exhibited small fluctuations about the 3-percent set point. To account for the random fluctuations in the oxygen concentration, the mean and the standard deviation (of heat of combustion) were obtained for a set of data points at several time intervals.

Experimental Procedure

The oxygen sensor was calibrated with mixtures of known oxygen concentrations so that a sensor output (in mV) versus oxygen-concentration calibration curve could be obtained. Subsequently, the system was flushed with air for several minutes to scavenge any fuel that was left inside the system from the previous experiment. Air from a pressurized cylinder was used for combustion, and its oxygen concentration was measured with the oxygen sensor prior to the experiment. The fuel flowmeter was also calibrated so that the number of counts/min from the optical encoder could be converted readily into flow rate in standard cubic centimeters per minute (scm).

A given air flow rate from the pressurized cylinder was initiated, and the fuel flow rate was gradually increased to a level at which a stable flame was established. The 3-percent oxygen concentration was dialed as a set point in the controller, and the controller was activated to maintain the fuel flow rate. The fuel flow-rate data in counts/min were recorded at regular intervals and were used in a correlation (described in the "Test Results" section) to obtain the value of heat of combustion. Over a time period, several data points were recorded, and the mean and the standard deviation were computed for the period.

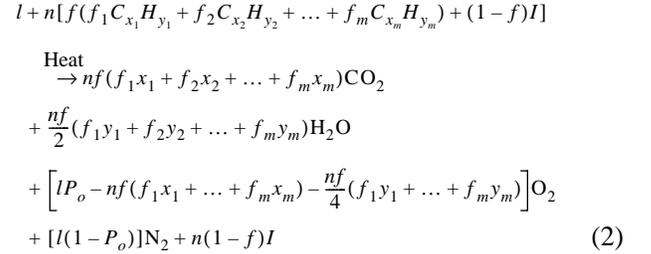
Theoretical Principles

The heat of combustion ΔH_{mix} of a gaseous mixture that has several saturated hydrocarbons and a noncombustible inert gas may be expressed as

$$\Delta H_{\text{mix}} = f \sum_{i=1}^m f_i \Delta H_i \quad (1)$$

where f is the total hydrocarbon mole fraction, m is the number of hydrocarbon species, f_i is the mole fraction of the i th hydrocarbon species ($\sum f_i = 1$), and ΔH_i is the heat of combustion of the i th hydrocarbon species.

The combustion of this mixture with air¹ may be expressed as



where l is the volumetric flow rate of air, n is the volumetric flow rate of the gaseous fuel mixture, I is the noncombustible inert fraction of the test gas, and P_o is the mole fraction of oxygen in the air.

The mole fraction of oxygen in the combustion products P can be expressed as

$$P = \frac{4lP_o - 4nf \sum_{i=1}^m f_i x_i - nf \sum_{i=1}^m f_i y_i}{nf \sum_{i=1}^m f_i y_i + 4n - 4nf + 4l} \quad (3)$$

Solving for f ,

$$f = \frac{4l(P_o - P) - 4nP}{n \left[4 \sum_{i=1}^m f_i x_i + (1+P) \sum_{i=1}^m f_i y_i - 4P \right]} \quad (4)$$

Substitution for f in equation (1) yields the following expression:

$$\Delta H_{\text{mix}} = \left[\frac{4l(P_o - P) - 4nP}{n} \right] \frac{\sum_{i=1}^m f_i \Delta H_i}{\left[4 \sum_{i=1}^m f_i x_i + (1+P) \sum_{i=1}^m f_i y_i - 4P \right]} \quad (5)$$

Let

$$A = 4 \sum_{i=1}^m f_i x_i + (1+P) \sum_{i=1}^m f_i y_i - 4P$$

A is a measure of the effective hydrocarbon content of the combustible component. Note that for saturated

¹Air is assumed to be composed of nitrogen and oxygen.

hydrocarbons, A can be further simplified since $y_i = 2x_i + 2$.

The ΔH_{mix} may now be expressed as

$$\Delta H_{\text{mix}} = \left[\frac{4l(P_o - P)}{n} - 4P \right] \sum_{i=1}^m \frac{f_i \Delta H_i}{A} \quad (6)$$

For a binary mixture of a single pure hydrocarbon species ($m = 1$) and an inert gas, the above equation reduces to

$$\Delta H_{\text{mix}} = \left[\frac{4l(P_o - P)}{n} - 4P \right] \frac{\Delta H}{A} \quad (7)$$

where $A = 4x + (1 + P)y - 4P$, and ΔH is the heat of combustion of the hydrocarbon species. Note that for methane ($y = 4$), A is independent of P .

For a mixture of unknown composition, the value of $\sum f_i \Delta H_i / A$ to be used in equation (6) is not known since f_i and A are unknown. Table I shows values of ΔH (ref. 8), A , and $\Delta H/A$ for various saturated hydrocarbons. (When more than one form of a hydrocarbon exists, such as n-butane and isobutane, the average value of ΔH is given.) A value of 0.03 for P is used in the calculation of A in each case. In the present technique, the adoption of a universal value of the $\sum f_i \Delta H_i / A$ coefficient in equation (6) is crucial for measuring the heat of combustion of a gaseous mixture of unknown composition. However, as noted from table I, $\Delta H/A$ varies from 25.94 for hexane to 26.6 for methane. For the present technique it is sufficient to adopt a value of this coefficient that will minimize the error in the prediction of the ΔH_{mix} for the domain of samples to be tested. Thus, for the domain of samples where all species occur with equal probability, the mean value of 26.27 is the appropriate choice for $\sum f_i \Delta H_i / A$ in equation (6).

In nature, however, hydrocarbons are not distributed uniformly. Methane makes up the bulk of the hydrocarbon component in most natural gas samples. Figure 3

Table I. Values of ΔH , A , and $\Delta H/A$ for Pure Saturated Hydrocarbons

Hydrocarbon	ΔH ,* kcal/mol	A ($P = 0.03$)	$\Delta H/A$
Methane, CH ₄	212.80	8	26.60
Ethane, C ₂ H ₆	372.82	14.06	26.51
Propane, C ₃ H ₈	530.61	20.12	26.37
Butane, C ₄ H ₁₀	686.65	26.18	26.23
Pentane, C ₅ H ₁₂	837.05	32.24	25.96
Hexane, C ₆ H ₁₄	993.51	38.30	25.94
			Mean = 26.27 ± 0.51 [†]

*All heats of combustion ΔH are given as the *gross* heat of combustion (producing liquid water at 25°C (ref. 8)).

[†]All errors in this paper are given at the 95-percent level of confidence.

shows the variation of $\sum f_i \Delta H_i$ with the methane mole fraction of the hydrocarbon component for 265 well samples that were analyzed in a 1982 Bureau of Mines survey of natural gases (ref. 9). Figure 4 shows the variation of $\sum f_i \Delta H_i / A$ for these samples. Although the value of $\sum f_i \Delta H_i$ varies significantly from sample to sample, the value of $\sum f_i \Delta H_i / A$ varies in a narrower range. Figure 5 presents a frequency distribution plot of $\sum f_i \Delta H_i / A$ for these samples. More than 90 percent of these well samples have a $\sum f_i \Delta H_i / A$ between 26.5 and 26.6. Thus, the mean value of 26.55 is the appropriate choice for $\sum f_i \Delta H_i / A$ for a wide range of natural gas samples.

Table II shows values of $\sum f_i \Delta H_i$, A , and $\sum f_i \Delta H_i / A$ for six natural gas pipeline samples from the Bureau of Mines survey. Although values of $\sum f_i \Delta H_i$ and A vary significantly from sample to sample, the value of $\sum f_i \Delta H_i / A$ is constricted to a narrow band from 26.53 to 26.59, in agreement with the mean value cited previously. Thus, although the adoption of a universal value for the $\sum f_i \Delta H_i / A$ coefficient inevitably introduces an error in the calculated value of ΔH , this error will be negligible in the overwhelming majority of samples from natural gas sources.

Table II. The $\sum f_i \Delta H_i$, A , and $\sum f_i \Delta H_i / A$ for Six Natural Gas Pipeline Samples

No.	Hydrocarbon component concentrations,* mole percent						$\sum f_i \Delta H_i$, kcal/mol	A ($P = 0.03$)	$\sum f_i \Delta H_i / A$
	CH ₄	C ₂ H ₆	C ₃ H ₈	C ₄ H ₁₀	C ₅ H ₁₂	C ₆ H ₁₄			
1	95.1	3.1	0.5	0.2	0.1	0.1	219.9	8.35	26.59
2	86.5	8.2	2.7	1.0	0.3	0.2	240.4	9.15	26.55
3	86.6	8.1	2.6	1.0	0.5	0.2	241.3	9.18	26.55
4	81.8	10.0	4.1	1.8	0.8	0.3	255.2	9.74	26.53
5	89.6	6.5	1.8	0.7	0.3	0.2	233.8	8.88	26.56
6	93.6	3.7	0.6	0.3	0.1	Trace	219.2	8.39	26.59
									Mean = 26.56 ± 0.04

*From reference 9.

Test Results

In the present study, $l = 7000$ sccm and $P = 0.03$. For these values equation (6) reduces to

$$\Delta H_{\text{mix}} = \left[\frac{28000(P_o - 0.03)}{n} - 0.12 \right] \sum_{i=1}^m \frac{f_i \Delta H_i}{A} \quad (8)$$

Equation (8) forms the basis for calculation of the ΔH_{mix} from the volumetric fuel flow rate measurements.

The present technique was first applied to two pure gas samples: methane and ethane. Since heats of combustion of these gases were already known, the results were used to determine the precision error of the method, and to this end a correction factor C_f was introduced in the calculation of volumetric flow rate from the optical encoder data E to correct for bias in the system. Table III shows the experimental data for methane that involves optical encoder data E in counts/3 min intervals, the fuel flow rate n in sccm calculated from the optical encoder data ($n = E/3 \text{ min} * 1000 \text{ cc}/1200 * C_f$), and the heat of combustion (ΔH) in kcal/mol calculated from equation (8); P_o was measured at 0.209. The mean value

Table III. Methane Data

E , counts/3 min	Fuel flow rate, n ,* sccm	ΔH , kcal/mol
2125	621.4	211.7
2118	619.3	212.4
2108	616.4	213.5
2115	618.4	212.7
2116	618.7	212.6
2108	616.4	213.5
2110	617.0	213.3
2118	619.3	212.4
2112	617.6	213.1
	Mean = 618.3 ± 3.1	Mean = 212.80 ± 1.22

* $C_f = 1.053$.

of the heat-of-combustion data is 212.80 ± 1.22 for methane.

Table IV shows the data for ethane; P_o was measured at 0.210. Readings were taken at 5-min intervals. The mean value of the heat-of-combustion data for ethane is 372.82 ± 2.60 . These two cases indicate that a precision approaching 0.6 percent can be achieved for pure saturated hydrocarbon fuels such as methane and ethane.

Table IV. Ethane Data

E , counts/5 min	Fuel flow rate, n ,* sccm	ΔH , kcal/mol
2122	356.2	370.9
2119	355.7	371.4
2121	356.0	371.1
2122	356.2	370.9
2117	355.4	371.8
2112	354.5	372.7
2113	354.7	372.5
2112	354.5	372.7
2107	353.7	373.5
2102	352.8	374.4
2108	353.8	373.4
2103	353.0	374.3
2103	353.0	374.3
2101	352.7	374.6
2104	353.2	374.1
	Mean = 354.4 ± 2.5	Mean = 372.82 ± 2.60

* $C_f = 1.007$.

The system was also used to find the heat of combustion of a mixture that was 92.49-percent methane and 7.51-percent nitrogen, which has a calculated heat of combustion (eq. (1)) of 196.82 kcal/mol; P_o was measured at 0.209. Table V shows data for this mixture, along with the fuel mole fraction f calculated from

Table V. Data for 92.49 Percent CH_4 Plus 7.51 Percent N_2

E , counts/3 min	Fuel flow rate, n ,* sccm	ΔH , kcal/mol	Fuel mole fraction, f (calculated from eq. (4))
2297	672.8	195.0	0.9162
2265	663.4	197.8	0.9294
2284	669.0	196.1	0.9215
2288	670.1	195.8	0.9199
2260	661.9	198.2	0.9315
2246	657.8	199.5	0.9374
2262	662.5	198.0	0.9306
2301	673.9	194.6	0.9146
2280	667.8	196.4	0.9231
	Mean = 666.6 ± 10.3	Mean = 196.82 ± 3.10	Mean = 0.9249 ± 0.0145

* $C_f = 1.054$.

Table VI. Summary of Data for Methane, Ethane, and Methane Plus Nitrogen

$$\Delta H_{\text{est}} = 48.6 + \frac{3.42 \times 10^5}{E}$$

Regression output: α , 48.5844; Standard error of ΔH_{est} , 2.005064; R^2 , 0.999463;
 Number of observations, 33; Degrees of freedom, 31; β , 342320.7; Standard error of β , 1425.077

Calibration gas	Flow data (E), counts/3 min	ΔH , kcal/mol	ΔH_{est} , kcal/mol
Ethane ↓	1061.0	372.82	371.22
	1059.5	372.82	371.68
	1060.5	372.82	371.38
	1061.0	372.82	371.22
	1058.5	372.82	371.99
	1056.0	372.82	372.75
	1056.5	372.82	372.60
	1056.0	372.82	372.75
	1053.5	372.82	373.52
	1051.0	372.82	374.29
	1054.0	372.82	373.37
	1051.5	372.82	374.14
	1051.5	372.82	374.14
	1050.5	372.82	374.45
1052.0	372.82	373.98	
Methane ↓	2125.0	212.80	209.68
	2118.0	212.80	210.21
	2108.0	212.80	210.98
	2115.0	212.80	210.44
	2116.0	212.80	210.36
	2108.0	212.80	210.98
	2110.0	212.80	210.82
	2118.0	212.80	210.21
2112.0	212.80	210.67	
Methane, 92.49 percent ↓	2297.0	196.82	197.61
	2265.0	196.82	199.72
	2284.0	196.82	198.46
	2288.0	196.82	198.20
	2260.0	196.82	200.05
	2246.0	196.82	201.00
	2262.0	196.82	199.92
	2301.0	196.82	197.35
	2280.0	196.82	198.73

equation (4). A value of 212.80 for ΔH and a value of 8 for A were used since methane was the only hydrocarbon involved. The mean value of the heat-of-combustion data is 196.82 ± 3.10 . The mean value of the fuel mole fraction data is 0.9249 ± 0.0145 .

Numerical Approach

The heat of combustion may also be expressed as

$$\Delta H = \alpha + \frac{\beta}{E} \quad (9)$$

where E is flow data, either in the form of flow units or directly in optical encoder counts, and α and β are constants determined from a least-squares regression analysis of flow data and heat of combustion for gases of various heat content, assuming constant values for l , P_o , and P . Equation (9) allows calibration of ΔH directly in terms of optical encoder counts using calorimetric gas standards, thus obviating the intermediate steps of quantifying the flow rate and oxygen concentration of the combustion air, calibrating fuel flow rate from optical encoder counts, and calibrating oxygen concentration from oxygen sensor voltage. This regression algorithm

was applied to optical encoder readings for the methane, ethane, and methane-plus-nitrogen data presented above and effectively provides a 3-point calibration of ΔH in terms of E (optical encoder data for ethane were normalized for 3-min intervals). Table VI summarizes these data and presents results of the regression analysis. A graphical plot of the data, the least-squares regression curve fit ΔH_{est} , and the ± 2 standard error of ΔH_{est} (SEE) band, which approximates the 95-percent confidence interval about the curve fit (ref. 10), are shown in figure 6.

Conclusions

An experimental technique has been developed and tested to measure the heat of combustion of a saturated hydrocarbon and inert gas mixture. The technique is especially attractive for applications in which an on-line measurement of the heat of combustion of natural gas is required. The system worked reasonably well during the course of laboratory investigation and thus validated the technique. Preliminary test results indicate that this technique can determine the heat of combustion of a gas of unknown composition with an uncertainty on the order of 2 percent.

NASA Langley Research Center
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January 22, 1996

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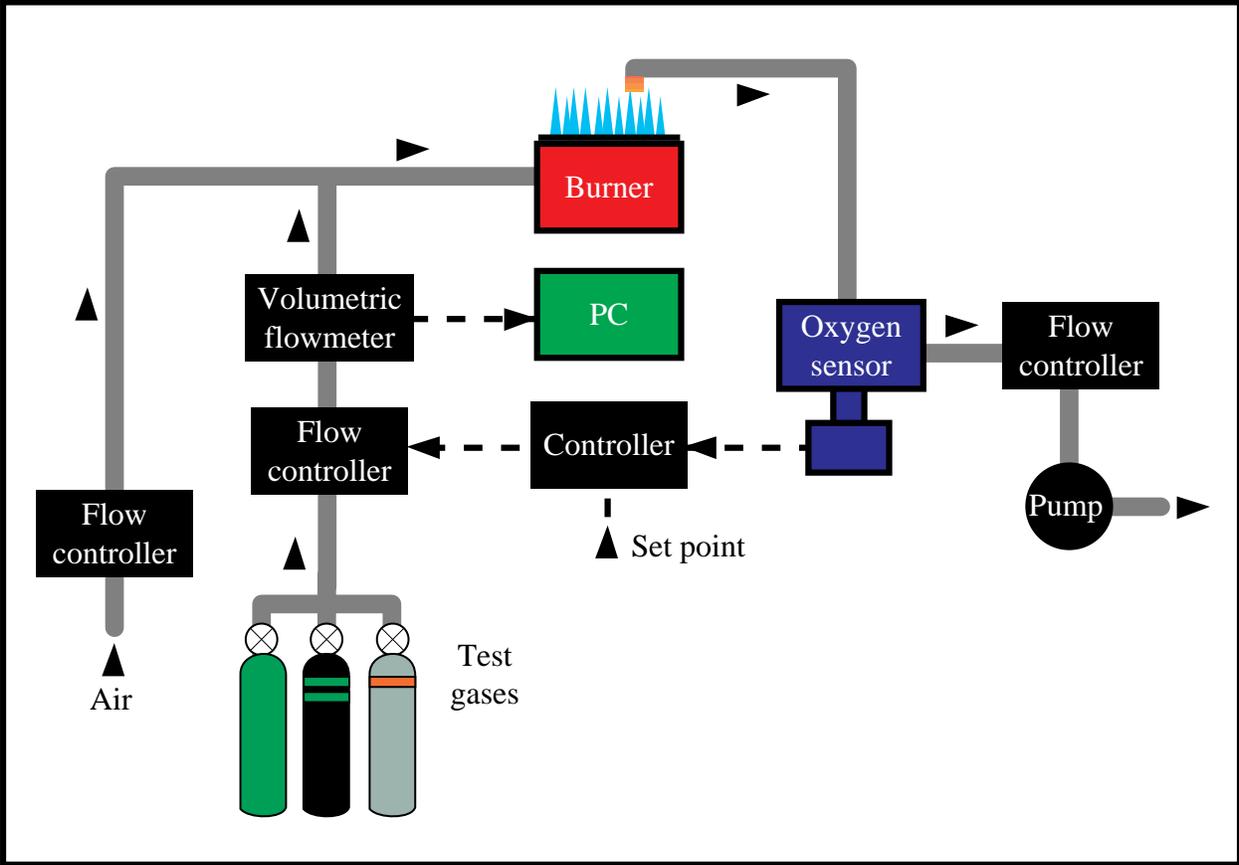


Figure 1. System diagram.



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Figure 2. Experimental setup.

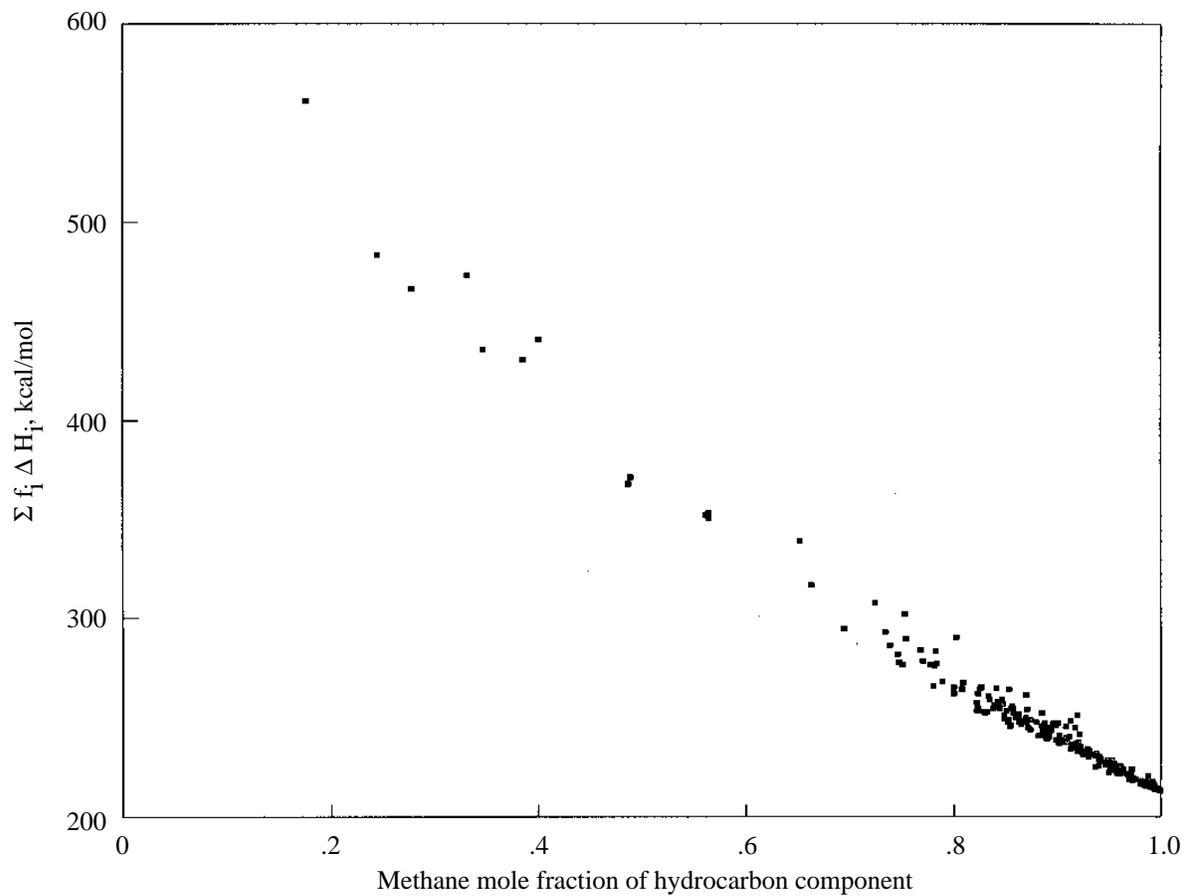


Figure 3. Variation in calculated heat of combustion $\Sigma f_i \Delta H_i$ with methane mole fraction of hydrocarbon component for 265 natural gas well samples.

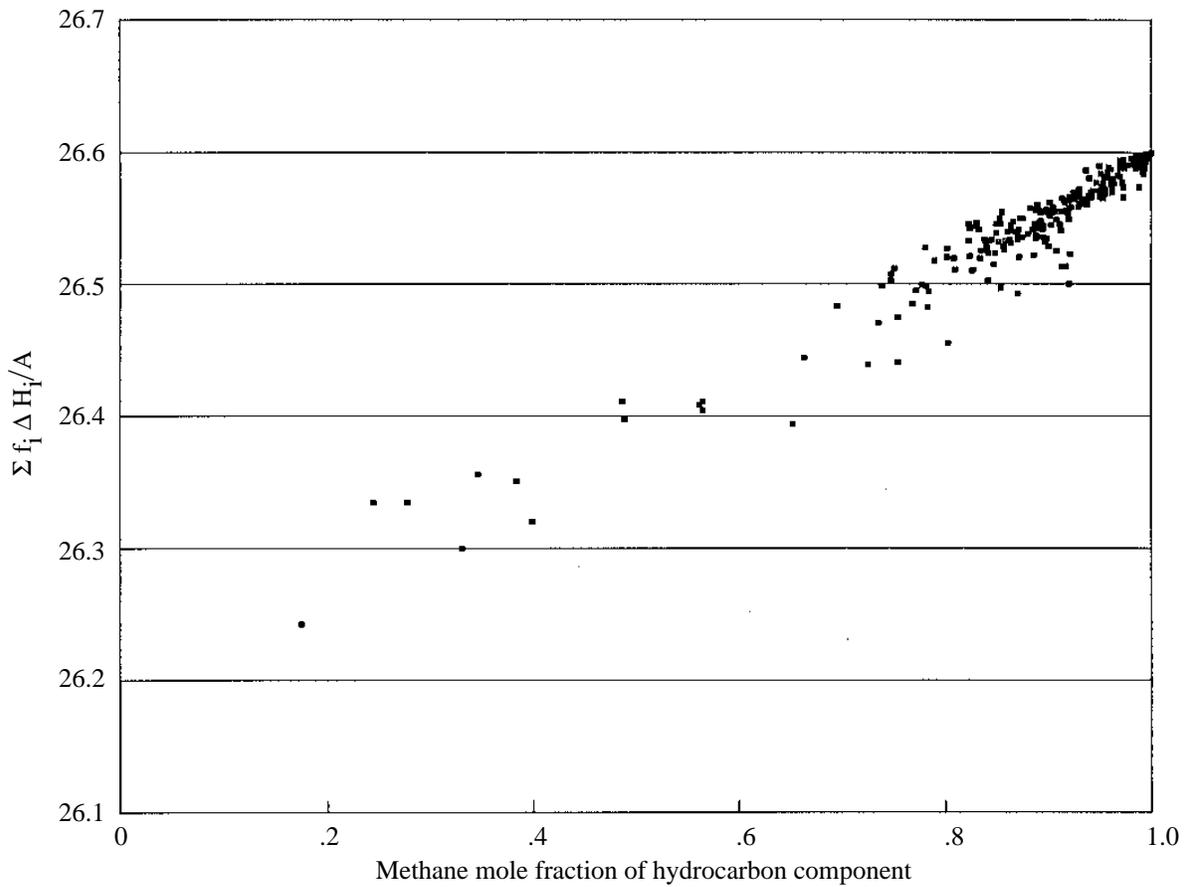


Figure 4. Variation of $\Sigma f_i \Delta H_i / A$ with methane mole fraction of hydrocarbon component for 265 natural gas well samples.

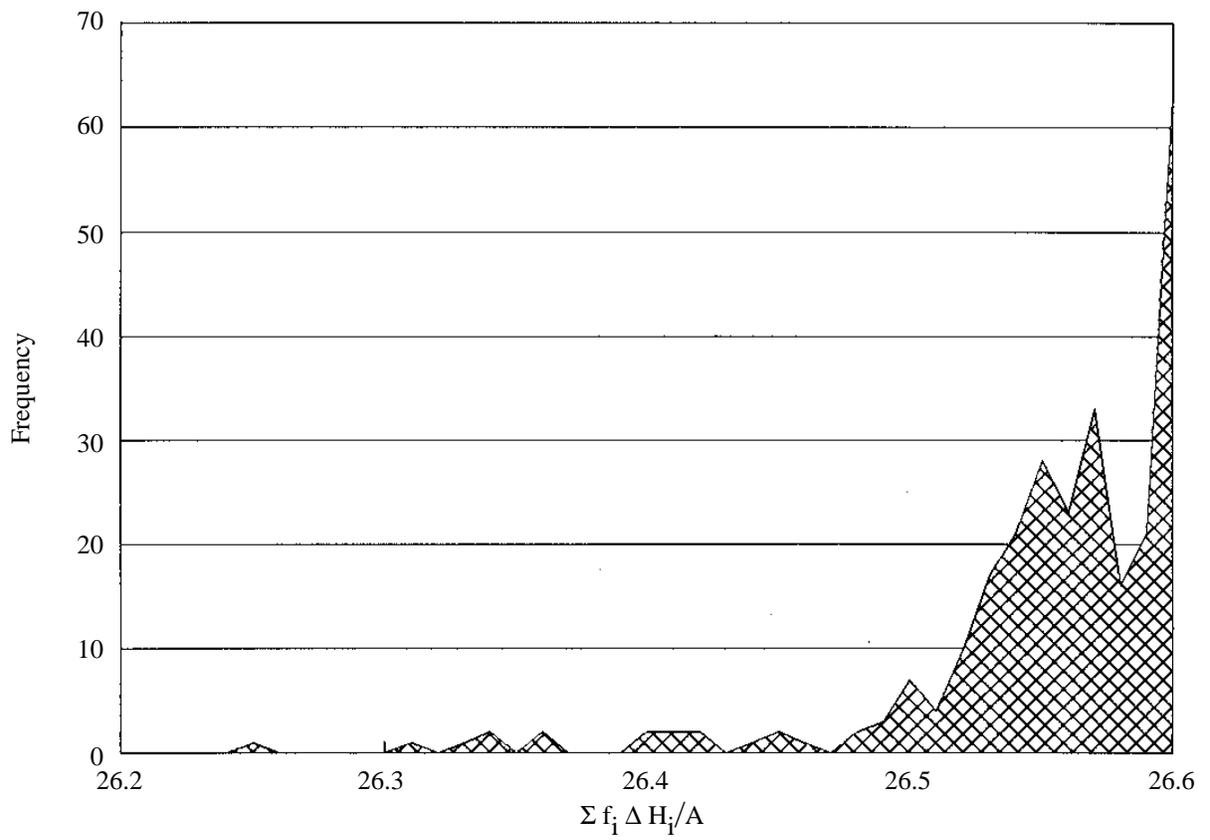


Figure 5. Distribution of $\Sigma f_i \Delta H_i / A$ for 265 natural gas well samples. Mean = 26.55; Standard deviation = 0.06.

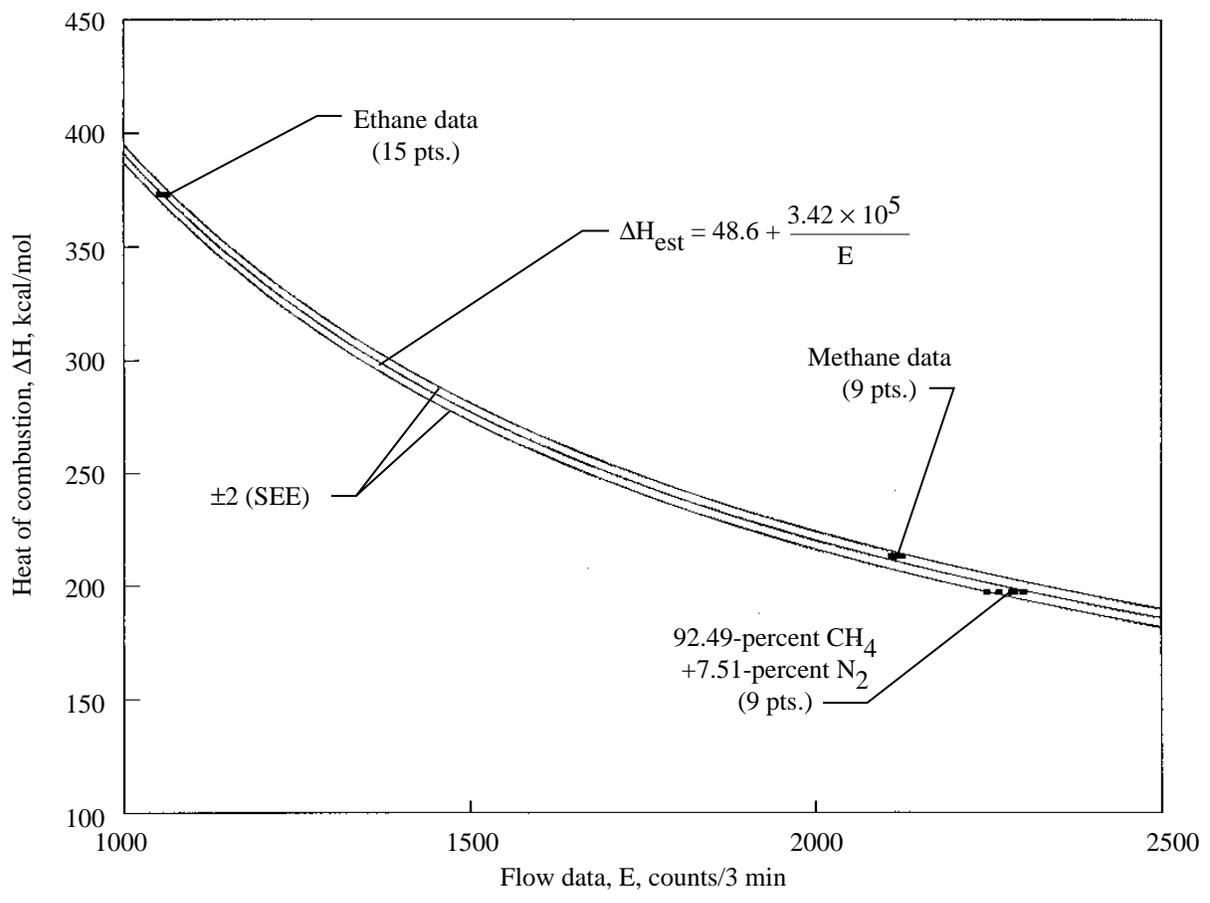


Figure 6. Summary of experimental data and regression results.

REPORT DOCUMENTATION PAGE

Form Approved
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1. AGENCY USE ONLY <i>(Leave blank)</i>	2. REPORT DATE March 1996	3. REPORT TYPE AND DATES COVERED Technical Paper	
4. TITLE AND SUBTITLE On-Line Measurement of Heat of Combustion of Gaseous Hydrocarbon Fuel Mixtures		5. FUNDING NUMBERS WU 763-01-41-31	
6. AUTHOR(S) Danny R. Sprinkle, Sushil K. Chaturvedi, and Ali Kheireddine			
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) NASA Langley Research Center Hampton, VA 23681-0001		8. PERFORMING ORGANIZATION REPORT NUMBER L-17511	
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) National Aeronautics and Space Administration Washington, DC 20546-0001		10. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA TP-3572	
11. SUPPLEMENTARY NOTES Sprinkle: Langley Research Center, Hampton, VA; Chaturvedi and Kheireddine: Old Dominion University, Norfolk, VA.			
12a. DISTRIBUTION/AVAILABILITY STATEMENT Unclassified-Unlimited Subject Category 35 Availability: NASA CASI (301) 621-0390		12b. DISTRIBUTION CODE	
13. ABSTRACT <i>(Maximum 200 words)</i> A method for the on-line measurement of the heat of combustion of gaseous hydrocarbon fuel mixtures has been developed and tested. The method involves combustion of a test gas with a measured quantity of air to achieve a preset concentration of oxygen in the combustion products. This method involves using a controller which maintains the fuel (gas) volumetric flow rate at a level consistent with the desired oxygen concentration in the combustion products. The heat of combustion is determined from a known correlation with the fuel flow rate. An on-line computer accesses the fuel flow data and displays the heat of combustion measurement at desired time intervals. This technique appears to be especially applicable for measuring heats of combustion of hydrocarbon mixtures of unknown composition such as natural gas.			
14. SUBJECT TERMS Natural gas; Natural gas energy content; Heat of combustion; Hydrocarbon combustion; Combustion physics; Combustion control; Enthalpy of combustion; Thermal energy measurement; Combustion calorimetry			15. NUMBER OF PAGES 13
			16. PRICE CODE A03
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT